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NUCLEAR DIVISION

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To (Name) H. F. Smith, Jr.
Division
Location Bldg. 9103, Y-12

Date September 23, 1977

Originating Dept.

Answering letter date

Copy to J. M. Case (2)
C. W. Holland
L. R. Phillips
R. D. Williams
P. E. Wilkinson
W. J. Yaggi
File - NoRC

Subject "Study on Y-12 MUF" Report

Enclosed is a draft of Study on Y-12 MUF, prepared in response to a request from Division of Classification, ERDA Headquarters.

A representative of the DOC, Headquarters, will be in Y-12 October 7, 1977 to discuss the study. It is felt desirable that the report be transmitted to ORO prior to the DOC visit; consequently, your comments are requested on an urgent basis. If no comments have been received by September 28th it will be assumed that the draft is satisfactory.

Document Y/LA-787, Report Explaining Historical Material Unaccounted for (MUF) (U) (Secret) was issued in January, 1977. Evidently DOC has not read that document; it will be retransmitted with the subject report.

Thank you for your cooperation.

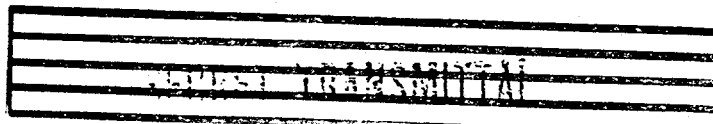
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NUCLEAR DIVISION

P. O. BOX Y, OAK RIDGE, TENNESSEE 37830

January 14, 1977

United States Energy Research and Development
Administration, Oak Ridge Operations
Attention: Mr. H. D. Hickman, Director
Manufacturing Division
Post Office Box E
Oak Ridge, Tennessee 37830

Gentlemen:

Report Explaining Historical Material Unaccounted For (MUF)

Attached is a MUF explanation report as requested in your letter of September 30, 1976, subject as above. The format has been modified from the outline enclosed with your requesting letter to facilitate presentation and explanation of the historical MUF.

If there are questions relative to the report, please call P. E. Wilkinson, Extension 3-5003.

Very truly yours,

J. M. Case
J. M. Case, Plant Manager
Oak Ridge Y-12 Plant

PEW:mld

Attachment - Y/LA-787

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No. 14 of 21 copies, Series A. 146

DOCUMENT NO. Y/LA-787

HISTORY OF MATERIAL UNACCOUNTED FOR (MUF)

I. INTRODUCTION

The Oak Ridge Y-12 Plant was built in 1943 and 1944 under the direction of the U. S. Army's Manhattan Engineer District. The initial purpose of the Y-12 Plant was the electromagnetic separation of U-235. In January 1947, the Atomic Energy Commission assumed control of the nation's nuclear energy effort, including the Y-12 Plant. Since that time, the electromagnetic method of separating U-235 has been discontinued. The Y-12 Plant has been assigned other problems and has become a sophisticated manufacturing and developmental engineering organization. The present principal responsibility of Y-12 is the production of components for nuclear weapons.

Early (electromagnetic) work at the Y-12 Plant was conducted under the pressures of World War II. At that time, because of the extreme shortage of material, major concern for uranium accountability was directed toward minimal losses and maximum utilization of available material. This report will not attempt to address accountability problems of that era.

Initial operations in the Y-12 Plant involving uranium enriched in U-235 used product from the electromagnetic separation process. By May 1947, production from the gaseous diffusion process completely replaced that from the electromagnetic process. Receipts from the gaseous diffusion process were in the form of UF₆. Until the latter part of 1947, product from the Y-12 Plant was in the form of UF₄. In 1948 uranium metal enriched in U-235 was first produced in Y-12. This was then used in the manufacture of uranium metal weapons components.

As in the case of any plant processing a material having value, systems have been set up to account for that material. The motivation for material accountability varies in relation to the value or strategic importance of the material being processed. Thus, in the case of a steel mill, the motivation is low when handling ore and increases through the manufacturing process. In the case of the Oak Ridge Y-12 Plant, material accountability relative to enriched uranium has historically been a major consideration. The basic approach has and continues to be that a good material balance is maintained within the Plant and within each processing area. Material balance may be expressed by a simple equation as follows:

Beginning Inventory + Receipts = Removals + Ending Inventory or (Beginning Inventory + Receipts - Removals) - Ending Inventory = Zero.

RESTRICTED DATA

This document contains Restricted Data as defined in the Atomic Energy Act of 1954. Its dissemination or disclosure to any unauthorized person is prohibited.

CLASSIFIED BY

L.R. Phillips

L. R. Phillips

Y-12 Classification Officer

1-17-77

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In practice using the second form of the equation, the zero is not obtained except by coincidence. The actual value obtained instead of zero has been variously termed Material Unaccounted For (MUF) or Book minus Physical Inventory Difference (BPID). Basically, MUF or BPID is, therefore, a measure of the lack of closure in the material balance equation. The lack of closure is equivalent to a variance account in fiscal accounting.

The objective of good accountability has been to provide an index of process control. The primary index has been the size of the monthly MUF. In an ideal situation the MUF would be zero. In an industrial or even a laboratory situation in real life the goal of zero MUF is not attainable. This may be illustrated by a simple analogy. If a glass is filled with milk to a specific mark, the milk is poured into a second glass and then into a third glass and then back into the first glass, it will not quite reach the original level. Some milk remains in both the second and third glass. This remainder is visible in the case of the milk. If the milk remaining in the second and third glass is not measured, it becomes Material Unaccounted For (MUF). Estimates can, of course, be made of the milk remaining in each glass and of any spillage or evaporation which might occur. These estimates when applied to the material balance would reduce the size of the MUF. However, care must be exercised not to overestimate these items or the glasses will become milk producers. A much more credible situation is to underestimate such amounts and have a larger positive MUF. This last process of underestimation has been the philosophy in general use at the Y-12 Plant in accounting for enriched uranium. One example will serve to illustrate this point. Prior to FY 1959 the amount of enriched uranium leaving the Plant in the storm sewers was generally estimated to be less than 10kg per year. In FY 1959, measurements showed 3.9kg per month was leaving via the storm sewers at that time. Prior to the FY 1959 measurements, the figure of 3.9kg per month would not have been credible. Subsequent additional attention to many of the details of the operations served to reduce the storm sewer losses.

As stated above, MUF has been used as a primary index of process control. The absolute value and variability of the MUF as a function of time have precluded its use as a primary means of detecting loss or diversion of enriched uranium. The primary system used to indicate loss or diversion has been and continues to be item accountability. In this system every item (piece, part, batch of material, discrete volume of solution, etc.) consisting of or containing enriched uranium is identified by a unique number. The item number and the quantity of enriched uranium in the item is entered in the perpetual inventory record. When any item loses its identity by chemical or physical change or by combination with other items, a new item number is assigned together with a new value

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for the contained amount of enriched uranium. During each physical inventory a complete listing is made of every item on hand. This listing is then compared to the perpetual inventory record and must agree perfectly so that each and every item is accounted for without exception. In addition, the measured or estimated amount of enriched uranium associated with the items is summed to determine the ending inventory for the material balance equation. Also, each MUF, regardless of sign or magnitude is evaluated by statistical methods to detect trends and to provide assurance that the MUF does not represent a real loss. If the item inventory agrees with the perpetual inventory record and the MUF obtained from the material balance equation is within expected limits, the conclusion may be drawn that no unaccounted for losses or diversions have occurred.

II. SUMMARY

An examination has been conducted of the MUF in the processing of uranium enriched to 20% or more in U-235 at the Oak Ridge Y-12 Plant. The thruput of U ($\geq 93\%$ U-235) in the Y-12 Plant is shown in Table I and Figures 1, 2 and 4. The thruput was defined as beginning inventory plus receipts in the casting area. The cumulative MUF as a function of time is also presented in Table I and has been plotted in Figures 3 and 4. The cumulative MUF is in terms of U enriched to 20% or more in U-235. It would appear from the graphs that two time intervals are of concern and should be separately examined. The first is the interval from FY 1958 through FY 1962. During this time, the cumulative MUF increased from about 60kg to about 470kg, an increase of 410kg. The second time period is from FY 1963 through FY 1976. During this latter time period, the cumulative MUF increased to a total of about 632kg, an increase of about 162kg. The relation between thruput and MUF is shown in Table I and in Figures 4 and 5.

Perusal of the history of the FY 1958 through FY 1962 time interval has led to the conclusion that the 410kg cumulative MUF associated with that time interval was primarily due to underestimating and understating the uranium content of discards, scrap equipment that was buried or salvaged and lack of adequate recognition of process losses in a large number of streams leaving the processing areas. This conclusion is discussed in detail below.

In a similar manner, information relative to the period FY 1963 through FY 1976 has been examined. Of the 162kg MUF associated with this period from 100kg to 120kg is attributable to the Super Kukla Program (20% enrichment) leaving a maximum of 62kg cumulative MUF for the time interval of fourteen years or about 4.4kg per year ($\sqrt{367}$ grams/month). This quantity is within the accuracy of the estimated and approximately measured discards and does not take into account accumulations in floors, walls, suspended

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ceilings, on equipment supports and inaccessible ventilation ducts such as
Detailed discussion of the conclusions relative to this period are presented below.

III. DISCUSSION

Early Enriched Uranium Processing

In the period from CY 1947 through FY 1954, the cumulative MUF for the Y-12 Plant showed a net gain of 12.4kg. This period

included recovery from a large number of Calutron components. This recovery operation resulted in the apparent gain in inventory.

Initial production of enriched uranium weapons components took place at the Y-12 Plant in the spring of 1948. Uranium hexafluoride (UF₆) from the gaseous diffusion process had been received in Y-12 since December 1946. By May 1947, all enriched uranium receipts in Y-12 were from the gaseous diffusion process.

The MUF for FY 1954 of material enriched to or above 20% was 7.7kg; in FY 1955 it was 17.6kg and in FY 1956 it was 26.2kg. During this time (FY 1954-FY 1956) many process changes were being made. No firm basis was available for estimation of process losses as the processes were still under development. Equipment holdups were not known.

Major clean out efforts indicated that holdup was underestimated. Estimates of holdup were modified from time to time, but retroactive corrections were not made since such corrections would have no real meaning relative to current performance. The MUF was considered an indicator of process control and of unmeasured process loss. In general such process losses of material enriched to or above 20% in U-235 were estimated or measured by means, which in retrospect appear crude, but which were essentially state-of-the-art measurements or best estimates based on available engineering judgment.

During this time the limit of error (LE) on the MUF was statistically determined by propagation of the inventory measurement LE's. If the MUF was within the LE, the process was considered to be in control. In about FY 1955 or FY 1956, propagation of LE's to determine an overall LE was abandoned due to

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MUF control limits were established by preparation of Shewnart-type control charts of MUF. The manufacturing system was considered to be in control if the MUF stayed within the control limits.

The FY 1958 Through FY 1962 Period (See Figure 3)

Several factors may be used to describe this period.

equipment was torn out and replaced, and new facilities were provided. 01d

During this period as a result of monitoring the MUF with control charts as previously described, several instances of "out-of-control" situations were noted. Such "out-of-control" situations resulted in extensive clean-out of equipment as well as committee evaluations. The net result of many such actions was to reduce the MUF but not to an acceptable level. The cumulative MUF is shown in Table I and Figure 3. As a consequence, a long-term committee, the Product Diversion Control Group, was set up to determine insofar as possible the location, course and magnitude of all streams of enriched uranium leaving the

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enriched processing areas. The committee found several previously unidentified streams and found that practically all previously identified streams had been underestimated by from 10% to very large factors. Again, no back corrections were made since they could not influence then current performance.

During this time period, the cumulative MUF increased by about 410kg as stated above. In addition, numerous distractions occurred which made it difficult to carefully assess the MUF situation. In addition to the large increase in thru-put, weapon designs were undergoing rapid development and evolution as were weapon delivery systems. As stated previously, extensive changes were being made in both the manufacturing and salvage processes with associated learning periods. Additional changes and learning periods were implemented after the nuclear criticality accident of June 1958. These included not only process changes but changes in methods of taking inventory. The cumulative effect of these distractions was to reduce the level of attention available for control of MUF.

The long-term committee, whose functions are defined above, was successful in identifying and measuring a large number of process losses which had previously been underestimated or not recognized as being worthy of consideration as measurable discards. Two examples of underestimated losses exemplify the findings of the committee. One was the loss to the storm sewer system which had been estimated at from 100 to 300 grams per month. This loss was computed by the committee based on actual measurements to be in the order of 3.9kg per month. A second example was the loss attributed to contaminated scrap metal and miscellaneous equipment of 13-15kg over a period of two and one-half years. The committee computed, based on extensive measurements, the actual loss to have been about 145kg for the same time period. A loss stream which had been considered minimal or inconsequential was the sanitary sewers. The committee at the time of their investigation found this stream contained over 270 grams per month.

Evaluating the results of the committee's findings and applying them to the available records of the FY 1957 through FY 1962 period where deemed appropriate leads to the conclusion that the actual process losses for the period were understated by from 350 to 400kg. Thus, from 10kg to 60kg must be explained by other means. It appears reasonable, based on historical data of the period, that losses which were estimated in addition to those evaluated by the committee could have been higher than the estimates and could account for the difference. Examples of such underestimation are: (1) the uranium content of heat-treating oil; (2) losses from dry chemistry (the systems for conversion of UF_6 to UF_4) and (3) unusually contaminated scrap. In explanation of the above three items, the following comments are applicable.

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Heat-treating oil was used during the production of thin-wall enriched uranium components. It was noted that oxide from the formed uranium components spalled from the uranium components into the oil during the heat-treating operation. Such spalling was not noticed during the earlier production period. When it was discovered, no attempt was made to estimate the uranium content of earlier discards of such oil to the burial ground. In addition, a number of drums which had been used to transport such oil were found to be contaminated, each containing a few tens of grams of enriched uranium.

Several occasions of losses from dry chemistry were noted. A radiation detector was standard equipment on the vent from dry chemistry. Although it was a state-of-the-art detector, it would be considered very crude by today's standards. Leaks from dry chemistry were detected either by alarms from the radiation detector or discovery of UO_2F_2 in the chemical traps on the system vent. Only rough estimates of the process loss prior to detection were possible.

In evaluating contaminated scrap and materials sent to burial, some batches were found to contain as much as 58 grams of enriched uranium per thousand pounds of scrap. The committee suggested a factor of 32.5 grams per thousand pounds of scrap. Prior to the committee factor of 32.5 grams/1,000 lbs, a factor of 10 grams/1,000 lbs had been used. It was noted that the contamination level of the scrap varied extensively. The 32.5 grams/1,000 lbs factor was a conservative value. More heavily contaminated scrap could easily have been discarded.

The FY 1963 Through FY 1976 Period

The period FY 1963 through FY 1976 has been characterized by several major changes in operations. During this period, the cumulative MUF increased about 225kg. These changes have been the cessation of UF_6 receipts from the gaseous diffusion cascades along with several special programs

The uranium inventory of the weapons system reached a level that permitted new weapons to be produced using the uranium from retired early weapons designs. It should be noted that enriched uranium from retired weapons is charged back into Y-12 at the original shipped value. This value assumed the components to be 100% metal. This may be a mistaken assumption. Two examples of this error are (1) plutonium containing primaries are disassembled at another plant and the enriched uranium decontaminated with respect to plutonium before shipment to the Y-12 Plant and (2) oxidation of components in stockpile may have resulted

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in oxide being abraded off and not returned to Y-12 for salvage. This latter also applies only to primary components. The overall impact of these two factors has not been quantitatively evaluated.

One special program was the manufacture of the Super Kukla fast burst reactor for the Lawrence Livermore Laboratory. The reactor consisted of a number of about 30 inch diameter circular plates of 20% enriched uranium metal alloy. Because of the physical size of the plates, they were cast in the foundry normally used for depleted uranium. The castings were machined as they were produced; the machine chips briquetted and used as part of a subsequent casting charge. During this time, it was also standard practice to briquette depleted uranium machine chips and use the briquettes as part of a casting charge. During the manufacture of the Super Kukla, a crossover of between 100kg and 120kg occurred. The inventory records were not corrected for this crossover. It was presumed to have occurred due to the mix-up between enriched and depleted briquetted machine chips. Assuming the crossover to have been 100kg, the balance of the cumulative MUF for the period is 125kg.

If the 125kg is averaged over the fourteen years, the average increase in cumulative MUF is 8.93kg per year. Based on 200 working days per year, this represents about 45 grams or about 1.6 ounces per day.

A second major program was the manufacture of Rover fuel elements for both the LASL and SNPO's contractor, Westinghouse. Rover fuel elements were made of extruded carbon loaded with uranium-containing beads. The extrusions contained different loadings of uranium as the development needs of the program changed.

A reactor core contained from 1,250 to 2,000 elements. In addition, nine out of every hundred coated elements were shipped for hot gas test to either the LASL or to Westinghouse. To produce one reactor core with the associated test elements required initiation of about 4,000 elements in the manufacturing cycle. During the manufacturing cycle, the uranium content of the blended material and the elements was checked at various stages of the manufacturing process. At least one major problem in the accountability system was documented. This referred to a technique used to sample Rover salvage batches. No quantitative evaluation of this problem was derived since such an evaluation would have been very speculative. Several other problem areas were noted. Among these were the following: Test elements after test were sawed into halves axially at both the LASL and Westinghouse. The halves were returned to Y-12 at the shipped uranium content, without credit for machine dust, during a part of the program. In addition, no accounting was made of losses during testing. Another problem area was that of evaluating the precise uranium content of a Rover element. The reactor designers were not primarily concerned with the precise uranium content, but rather with

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the relative content along the length of an element and the relative content between elements. Variations of total uranium content, within rather broad limits, could be accommodated by the reactor control system. As a consequence, primary emphasis was placed on providing information required for reactor design with accountability information having somewhat lower priority. The gamma counting techniques used to measure fuel element uranium content were the best available and were continuously compared to standards. Preparation of the standards and their evaluation was done using the best available techniques. [No firm estimate of bi

Much of the uranium in reject fuel elements was recovered by private enterprise. The uranium in irradiated fuel elements is still being recovered. As a consequence, the overall material balance loop has not been closed, and no final evaluation at the Y-12 Plant can presently be undertaken.

In a qualitative evaluation of the relationship between the Rover MUF and the Y-12 MUF, it is evident that the Rover Program contributed to the increase of the Y-12 MUF. However, because of the difficulties previously noted in the Rover Program with the attendant measurement uncertainties as well as the variable time relationships between manufacture and scrap recovery, it has not been possible to quantitatively evaluate the relationship between Rover MUF and Y-12 Plant MUF.

IV. CONCLUSIONS

As stated previously, the general philosophy followed by the Y-12 Plant in estimating discards and in stating the value of measured discards of very low concentration or inadvertant discards has been to be conservative. This philosophy has been in consonance with the use of MUF as a measure of process control. In addition, this has resulted in generally defensible and credible values. As an example, no allowance has been made for accumulations of material [nor has any allowance been made for material tracked out by miscellaneous equipment passing through areas nor for track-out by personnel not normally working in enriched processing areas. Allowance has not been made for holdup in building structures. This in itself could contribute significantly to the MUF as evidenced by the silver recovered during and subsequent to the demolition of the old San Francisco Mint.

During this time frame, the factors used to describe uranium discards or removals by various means have been evaluated on a continuing basis. It

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should be recognized that such items as "stack losses," although measured using the best available technology, are only best estimates of the true value. Similar comments apply to storm sewer measurements, track-out, contaminated scrap metal, laundry water, sanitary sewers, etc. Under these circumstances an apparent MUF of about 1½ ounces per day could readily result from a small underevaluation of the listed discard streams.

Calculation of Corrected MUF

A quantitative evaluation of the reported MUF and a corrected MUF is presented below. The calculations are based on the assumption that discards as described by the Product Diversion Control Group FY 1957 through FY 1960 period. Subsequent to this period, the major recommendations of the Product Diversion Control Group would have been implemented. The value of 150.2kg used for FY 1959 is taken directly from the Product Diversion Control Group Interim Summary Report, Y-B65-302, dated September 21, 1959.

Calculation

Total

399.4kg Correction

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Cumulative MUF through FY 1962	470.3kg
Correction	<u>-399.4kg</u>
Unexplained MUF through FY 1962	70.9kg
Cumulative MUF through FY 1976	632.3kg
Cumulative MUF through FY 1962	<u>-470.3kg</u>
Cumulative MUF FY 1963 through FY 1976	162.0kg
Super Kukla Crossover	<u>-100.0kg</u>
Unexplained MUF	62.0kg
Total Unexplained MUF	70.9kg (from Period through FY 1962)
	<u>+62.0kg (from Period through FY 1976)</u>
Total	132.9kg
Total Unexplained MUF	133.0kg

It would appear reasonable to attribute this unexplained MUF to the MUF due to Rover, holdup in structures undetermined biases in measurements and estimated values.

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